ChinaXiv合作期刊

Nuclear Science and Techniques **24** (2013) 030301

Interaction between uranium and humic acid (II): complexation, precipitation and migration behavior of U(VI) in the presence of humic substances

LIAO Jiali[1] WEN Wei[1,2] LI Bing[1] YANG Yuanyou[1] ZHANG Dong[2]
KANG Houjun[2] YANG Yong[2] JIN Jiannan[1] LIU Ning[1,*]

[1]*Key Laboratory of Radiation Physics and Technology, Ministry of Education, Institute of Nuclear Science and Technology, Sichuan University, Chengdu 610064, China*

[2]*Institute of Nuclear Physics and Chemistry, CAEP, Mianyang 621900, China*

Abstract The complexation, precipitation, and migration behavior of uranium in the presence of humic acid (HA) or fulvic acid (FA) were investigated by cation exchange, ultrafiltration and dynamic experiment, respectively. The results showed that (i) complex equilibrium between the uranium and humic substances was achieved at approximately 72 h, (ii) the coordination number varied from 1:1 to 1:2 (U(VI) : humic acid) as pH increased from 3 to 6; and (iii) , while the complex stability constant decreased when temperature increased, but increased with pH value. We found that the precipitation of uranyl could only be observed in presence of HA, and the precipitation was influenced by conditions, such as pH, uranium concentration, temperature, and the HA concentration. The maximum precipitation proportion up to 60% could be achieved in the condition of 40 mg/L HA solution at pH 6. We further observed that the migration behavior of uranium in soil in the presence of humic acid (HA) or fulvic acid (FA) was different from that in the presence of inorganic colloid, and the effect of humic substances (HS) was limited.

Key words Interaction, Uranium, Humic acid, Complexation, Migration, Precipitation, Soil

chinaXiv:202306.00189v1

1 Introduction

Radioactive wastes or contamination were generated inevitably along with the development of nuclear weapons, nuclear power, and other nuclear techniques. Usually, for low-medium level or very low level radioactivity, waste materials were stored in a shallow underground waste repository in-situ with a multiple barrier system. Unfortunately, the radioactive waste would release into environment if the barriers were damaged or lose efficacy. Additionally, wastes from running nuclear facilities and ex-service equipments could directly release into environment. For decades, safe treatment of radioactive wastes has been a great concern all over the world and still is in a continuously developing stage[1-5].

The investigation of the precipitation and migration behavior of radionuclides in the environment is vital for the evaluation of the trace of radionuclides and the safety assessments of underground repositories. As an important actinide, uranium could react easily with natural media including inorganic and organic matter in the environment due to its active chemical properties. This high reaction activity influences the chemical species and behavior of uranium, and further influences the migration behavior of uranium in environment[6-8]. Thus, the investigation of interaction between uranium and the natural media, especially with natural organic matter, would help us better understand the transportation of actinides in natural environment.

Among the so many natural substances, HS, which accounts for more than 50% of the natural

Supported by China National Natural Science Foundation (Nos.21071102 and 91126013), Joint Funds of China National Natural Science Foundation and China Academy of Engineering Physics (No. 10476015) and National Fund of China for Fostering Talents in Basic Science (No.J1210004)

* Corresponding author. *E-mail address:* nliu720@scu.edu.cn

Received date: 2010-09-17

ChinaXiv合作期刊

chinaXiv:202306.00189v1

organic materials and are widely distributed in soils, water, and precipitates of the ecosphere, are divided into FA, HA, and humin according to their molecular weight or solubility in different acidic solution. As we know, FA consists of moderate-molecular-weight organic substances of non-specific composition and soluble at all pH values; HA consists of high-molecular-weight organic substances and is soluble in alkaline media (e.g. in 0.1mol/L NaOH), but insoluble in acidic media (at pH 1~2); humin is composed of portions of organic matter in soils or sediments and is insoluble at any pH value[8,9]. Among these substances, FA and HA would actively interact with many metal ions, such as Cu (II), Ni (II), Co (II), Np (V), Am (III), Eu (III) and Cm (III) *via* adsorption[10], complexation[11-17] or redox reaction[18] by their functional groups, such as carboxyl (-COOH), carboxide (-CO), methoxy (-OCH_3), and hydroxyl (-OH).

The HS (including FA and HA) in underground water, sediment or synthesized complexation with rare metal, heavy metal, as well as actinides has been studied widely using electrophoretic ion focusing[19,20], ion exchange[21], UV spectroscopy[22], time resolved laser fluorescence spectroscopy (RTLFS)[23], ultrafiltration, and so on[17]. The species of metal ion were also studied using mathematical model in the presence of HS[24,25]. The complexation of natural humic substance colloid with metal ions in raw natural environment has been determined directly[26-29].

The results from the above mentioned experiments implied that complexation is an important factor that influences the migration or precipitation behavior of high valent metal ions besides adsorption and oxidation-reduction[6-8,25,30,31]. Therefore, the investigation of interaction between uranium and HS is significantly important for understanding the chemical behavior and species of uranium in soils or underground water, which will not only help us to predict the transport of uranium in natural environment but also better guide the safety evaluation for nuclear waste disposal.

In our previous paper, the adsorption of uranium on HA solid has been discussed[32]. The experimental results showed that uranium could be adsorbed on solid HA under some conditions, and the adsorption was influenced by pH, temperature, origin of humic acid as well as other factors[32]. In this paper, in order to further understand the interaction between uranium and HA, the complexation of HS with uranium was carried out with Schubert method, and the precipitation and migration of uranium were performed.

2 Experimental section

2.1 Materials

Soil samples[32] were dried at room temperature, ground, sieved to 2.0 mm after removal of plant roots, and sealed in polyethylene bags for use. Its components are as same as that in literature [33]. Stock solution of U(VI) was prepared by uranyl nitrate ($(UO_2NO_3)_2.6H_2O$), and its concentration was calibrated by mass analysis. Other chemical reagents were of analytical grade unless stated otherwise. The glassware were rinsed by distilled water and allowed to dry before use.

X-5 macroporous adsorption resin (surface area: 500–600 m^2/g; average pore diameter: 29–30 nm) purchased from The Chemistry Plant of Nankai University was refluxed with hot ethanol following washed with cool ethanol, then kept in ethanol for use. 001×7×7 strong acidic cation ion exchange resin was purchased and pretreated with HCl and NaOH in sequence to transform it from H^+ to Na^+ form before use.

The centrifuge tube with a molecular weight cutoff of 3 KD was obtained from Millipore Company. The 0.45 μm of microfiltration membrane was bought from Shanghai Xinya Device Factory. Migration column (Φ10×150 mm) was custom made with plexiglas in our laboratory.

2.2 Measurement of U(VI)

The concentration of U(VI) in solution was determined directly by UV/Vis spectroscopy or Anodic Stripping Voltammetry. U(VI) in soil column was determined after transformed into aqueous solution. The methods are described as below.

ChinaXiv合作期刊

The trial solution of 10 mL digested by 0.8 mL of 30% H_2O_2 and 0.2 mL of 16 mol/L nitric acid was washed with 5 mL buffer solution (chloroacetic acid-sodium acetate, pH = 2.5) into 25 mL volumetric flask. Then, the final volume was increased to 25 mL with 1 mL of 0.6% arsenazo (Ⅲ) aqueous solution and distillation water for determination by UV/Vis spectroscopy.

The soil sample was weighed, put in a vessel, and digested with hot HCl (10 mL), HNO_3 (5 mL) and H_2SO_4 (1 mL). After the sample was evaporated entirely, 20 mL of 40% HNO_3 was added to dissolve the residual salt. The solution was then filtered by neutral filter paper, and the filter solution was separated with TBP extraction resin chromatographic column. Finally, together with 2 mL mixture solution (25 g 1,2-Cyclohexanedia minetetraacetic acid, 5 g NaF and 65 g sulfosalicylic acid were dissolved in 1 L of pH 8 NaOH solution), eluted uranium solution was adjusted to a pH value of 7.

For very low concentration uranium($<1\,\mu$g/L), digestive trial solution was mixed in chloranilic acid solution and assayed by Anodic Stripping Voltammetry.

2.3 Complexation experiment by cation ion exchange method

This method is based on retention of the polycation humate complex on the cation exchange resin while the non-complexed uranyl cation passes through[21]. A series of batch cation ion exchange experiments were performed to calculate the stability constant of UO_2^{2+} with HS.

All experiments were conducted at a total uranium concentration of 1.0 mg/L, the mass of resin in each sample was 0.5 g at pH range from 3 to 6. Ion strength of 0.10 was maintained using $NaClO_4$ solution and pH was adjusted with dilute NaOH or HCl. The concentrations of HA and FA were varied from 5 to 10 ppm and 10 to 200 ppm, respectively. All experiment samples were kept in 30 mL vessel and shaken continuously for 96 h at the steady state temperature. The aqueous U(VI) was then determined using UV/Vis Spectroscopy or Anodic Stripping Voltammetry as described above.

2.4 Precipitation experiment

Ultrafiltration was used to separate the free uranyl ion from the uranium-HS precipitate. The free uranyl ion and micro-humic matter could pass through the filtration membrane, while the complex humate, macro-HC as well as HA which adsorbed U(VI) were retained on the membrane. Based on orthogonal method, which was used to determine the optimum reaction condition, the further experiment was performed as following:

20 mL of 10 mg/L uranyl nitrate solution was added in 50 mL vessel that contained HS and electrolyte solution. The required concentrations of HS and electrolyte were then regulated and the vessel was shaked. After shaking for 24 h, the reaction solution was filtered *via* 0.45 μm microfiltration membrane. The residual U(VI) in the filtrate was tested and the precipitation ratio was calculated using the following formula:

$$P\% = (C_0 - C) / C_0 \times 100\% \qquad (1)$$

where C_0 is original concentration of UO_2^{2+} (mg/L), C is residue concentration of UO_2^{2+} (mg/L), P% is the percent of U(VI) precipitate.

2.5 Migration experiment

Migration experiments were carried out in plexiglas column (Φ10× 150 mm) and pumped at a rate of 12 mL/d with the groundwater (spray column every 6 hours at about 2.8 mL/h) in order to keep the water flow velocity in the experimental columns being close to the natural groundwater flow velocity. The characteristics of the soil sample are detailed in Table 1. After 600 days, the experimental columns were dismounted, then placed for several days to remove the extra water in the column. Then the columns were cut into 2-cm thick slices, the concentration of UO_2^{2+} in soil was tested as described above.

The flow rate of groundwater in the experimental column was determined by breakthrough curve of HTO and the ^3H in the outflow was analyzed with a liquid scintillation analyzer.

The migration results were expressed as the distribution coefficient (Kd), the retardation factor (Rd), and the maximum migration distance (d, mm):

chinaXiv:202306.00189v1

ChinaXiv合作期刊

$$R_d = \upsilon_w / \upsilon_n \quad (2)$$

$$K_d = (R_d - 1) \times \varepsilon / \rho \quad (3)$$

where υ_w is the flow velocity of groundwater(cm/d), υ_n is the migration rate of U(VI) (cm/d), ε is the effective porosity of the soil, and ρ is the bulk density of the soil (g/ cm^3).

Table 1 Columns and soil samples for uranium migration experiment

Columns	Soil samples	Elution water	Soil densities / g·cm^{-3}
1	Soil only	Ground water	1.08
2	Soil	Ground water containing 1g/L FA	1.06
3	Soil:PHA (10:1.5)	———	1.04
4	Soil:EHA (10: 1.5	———	1.04
5	Soil: $Fe(OH)_3$ (10g :20mL of 87% $Fe(OH)_3$ solution)	Ground water	1.06
6	Soil: montornollite(10:2)	———	0.98

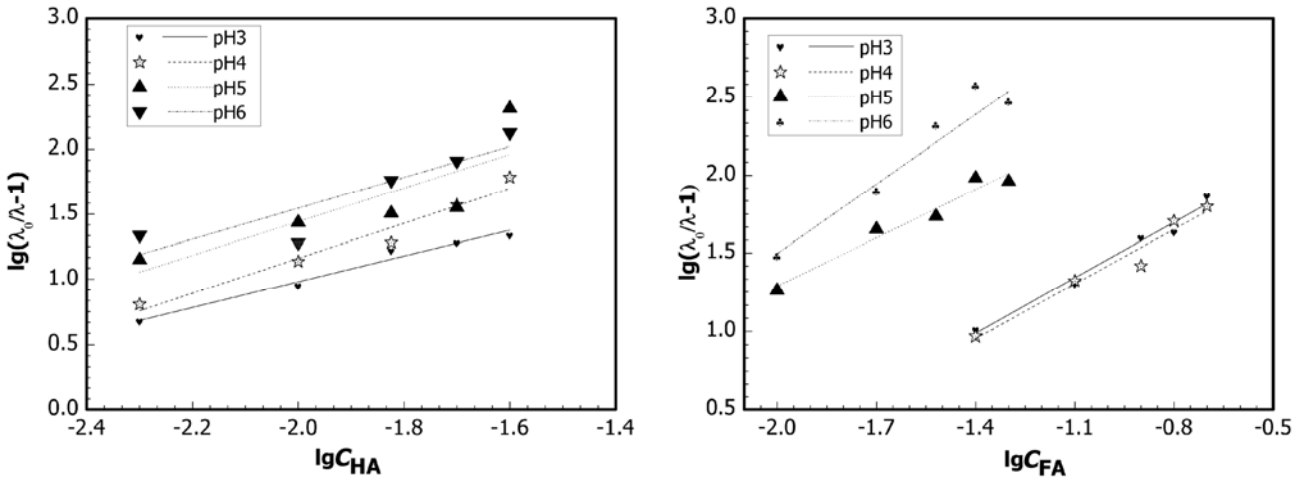

Fig.1 Effect of pH on complexation of UO_2^{2+} with HA and FA C_0 (UO_2^{2+})=1 mg/L–0.1 mol/L$NaClO_4$, t=96 h, T=25℃, m_{resin}=0.5 g Fitlinearcurve fomula were displayed as following: HA: y_{pH3}=0.99x+2.96, r^2=0.9777; y_{pH4}=1.35x+3.85, r^2=0.9586; y_{pH5}=1.29x+4.53, r^2=0.6776; y_{pH6}=1.19x+4.40, r^2=0.8073. FA: y_{pH3}=1.19x+2.65, r^2=0.9763; y_{pH4}=1.18x+2.60, r^2=0.9700; y_{pH5}=1.14x+3.79, r^2=0.9207; y_{pH6}=2.81x+6.06, r^2=1.0000.

3 Results and discussion

3.1 Complexation of U with HS

3.1.1 Equilibrium time

Equilibrium time of the complexation of UO_2^{2+} with HA or FA was obtained through a time-study experiment. Our results implied that this multicomponent system equilibrium occurred in two actions, including (i) ion exchange with cation exchange resin and (ii) complexation of U(VI) with HA or FA. During the first 32 h, ion exchange was a dominant process which resulted in a decrease of UO_2^{2+} in aqueous solution along with time. After 32 h, complexation became dominant and we observed an increase of UO_2^{2+} in aqueous solution until the increasing trend plateau approximate at 72 h. So 96 h was selected as the equilibrium time in experiment for complexation of UO_2^{2+} with HA or FA entirely.

3.1.2 Effect of pH on complexation of UO_2^{2+} with HA and FA

The effect of pH on complexation was performed in the range of pH 3 to pH 6 and the result is shown in Fig.1. The stability constant of complexation increased with the increase of pH value from 3.0 to 6.0. This trend was most likely caused by release of H^+ from function group such as carboxyl, hydroxyl or others, which consequently attributed to the combination of UO_2^{2+} with HS at high pH value. On the contrary, the dissociation of HS was retarded by H^+ concentration at low pH value, so that the complexation of UO_2^{2+} with HS was slowed down in aqueous solution. We

chinaXiv:202306.00189v1

ChinaXiv合作期刊

found that the complexation tendency of HA was more distinguished than that of FA (Fig.1).

3.1.3 Effect of temperature on complexation

Experimental results of complexation with HA and FA at different temperature are shown in Fig.2. The stability constant in the range from 10℃ to 20℃ was clearly lower than that at high temperature from 30℃ to 40℃. In the range from 0℃ to 20℃, complexation stability decreased as temperature increased. But at the range from 30℃ to 40℃, complexation stability increased when temperature increased. The reason might be that HA/FA complexed with uranium in its essential state at low temperature less than 20℃, but polymerized with each other before the reaction of complexation at high temperature from 30℃ up to 40℃. We believed that the polymerization helps to generate more stable complex compound. The stability constant of HA is always higher than that of FA in the same condition which further supported our speculation.

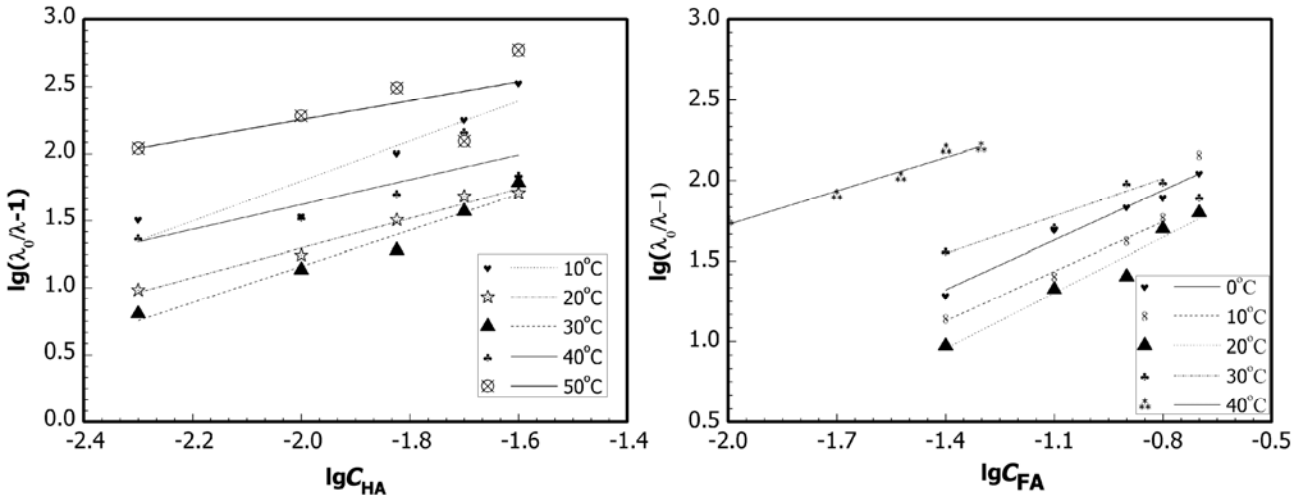

Fig.2 Effect of temperature on complexation of UO_2^{2+} with HS. C_0 (UO_2^{2+})=1mg/L–0.1mol/L$NaClO_4$, t=96 h, T=25℃, m_{resin}=0.5 g. Fitlinearcurve fomula were displayed as following: HA: $y_{0℃}$=1.50x+4.81, r^2=0.8588; $y_{10℃}$=1.10x+3.50, r^2=0.9806; $y_{20℃}$=1.35x+3.85, r^2=0.9586; $y_{30℃}$=0.71x+2.03, r^2=0.8621. FA: $y_{0℃}$=1.03x+2.77, r^2=0.9771; $y_{10℃}$=1.04x+2.59, r^2=0.9921; $y_{20℃}$=1.17x+2.59, r^2=0.9638; $y_{30℃}$=0.98x+2.88, r^2=0.8950; $y_{40℃}$=0.70x+3.12, r^2=0.9690.

Table 2 Factors and level

Factors		Level		
		1	2	3
pH		4	6	8
Concentration of HS, mg/L	HA	0	50	100
	FA	0	50	100
Time, d		1	2	4
Ion strength, 0.1mol/L		0.01	0.1	0.5
Concentration of UO_2^{2}, mg/L		1	5	10
Coexist ion		HCO_3^-	CO_3^{2-}	SO_4^{2-}

Note: HCO_3^- in acidic solution (at low pH); CO_3^{2-} in alkaline solution (at high pH).

3.2 Precipitation of uranium

3.2.1 Determination of influence factors for precipitation experiment

The choice of influence factor on precipitate of uranium in the condition of humic substance was performed with orthogonal experiment. In this experiment, $L_{27}(3^{13})$ schedule was chosen for 7 factors and 3 levels (Table 2). The orthogonal experiment results showed the effect factors on precipitation in the presence of HA were in degressive order as following: pH, concentration of uranium, temperature as well as concentration. But in the presence of FA, only temperature could influence the precipitation. Based

chinaXiv:202306.00189v1

ChinaXiv合作期刊

on the orthogonal experiment, the influencing factors used for the condition experiments were determined.

3.2.2 Effect of pH on precipitation

Precipitation experiment was performed in 10 mg/L of uranium and 0.1 mol/L of $NaClO_4$ mixture solution. pH influenced precipitation of U(VI) efficiently in HA but indistinctly in FA as described in Fig.3. This result was consistent with that of orthogonal experiment. Furthermore, the percent of precipitate increased with the increase of pH in the range of pH 3 to pH 6, but decreased with the increase from pH 6 to pH 9. In other words, about 65% of uranyl humate would be retarded on 0.45 μm micropore membrane at pH 6 that was close to the environment pH, but with pH value increasing or decreasing from pH 6, the precipitate would dissolve gradually. However, uranyl humate passed through the membrane at any pH value.

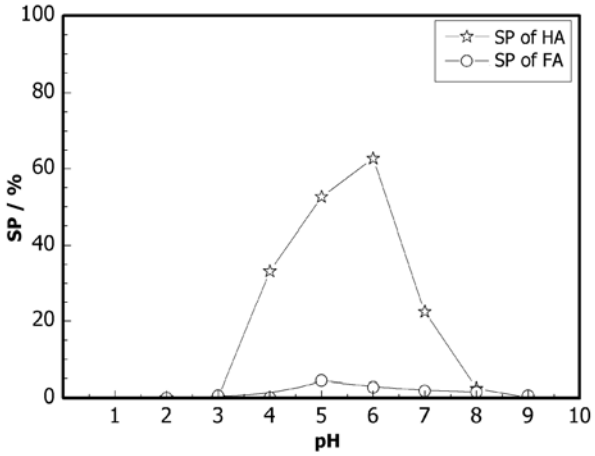

Fig.3 Effect of pH on precipitate of UO_2^{2+}. C_U=10 mg/L, C_{HA}=40 mg/L, C_{FA}=100 mg/L, I=0.1 mol/L and T=30℃.

3.2.3 Effect of uranium concentration

The effect of the concentration of uranyl humate on precipitate was secondary to that of pH. The result was plotted in Fig.4. The curve implied that the precipitate was uranium concentration dependent. When uranium concentration was lower than 5 mg/L, the percent of precipitate increased along with the increase of uranium concentration; however, in the range from 5 mg/L to 100 mg/L, the percent of precipitate decreased along with the increase of uranium concentration.

It was likely that the sample solution that maintained approximately 40 mg/L HA limited the complexation/adsorption capacity for uranium. As a result, the maximum precipitate (approximate 60%) took place at uranium concentration of 5 mg/L.

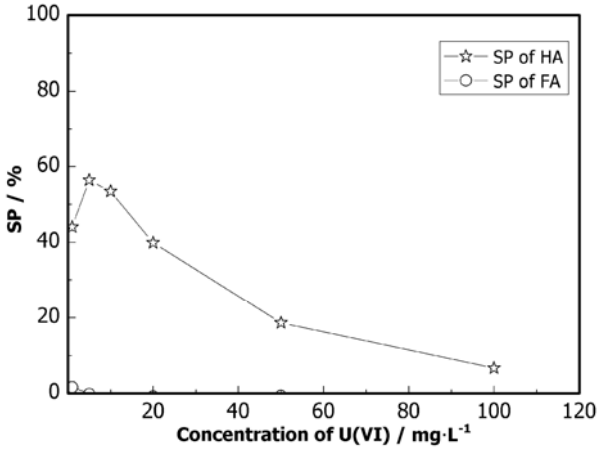

Fig.4 Effect of concentration of UO_2^{2+}. C_{HA}=40 mg/L, C_{FA}=100 mg/L, pH=6, I=0.1 mol/L and T=30℃.

3.2.4 Effect of HA concentration

The test was carried out in the condition of 10 mg/L uranium solution at pH 5. The result was shown in Fig.5. Based on this figure, apparent effect of HA concentration on uranyl humate was observed. But the effect was limited in the condition of pH 5, uranium concentration of 10 mg/L, as well as mixing for 1d. With the concentration increasing to 40 mg/L, the percent of precipitate varied no longer at maximum precipitate of approximate 60% which was according with the data above.

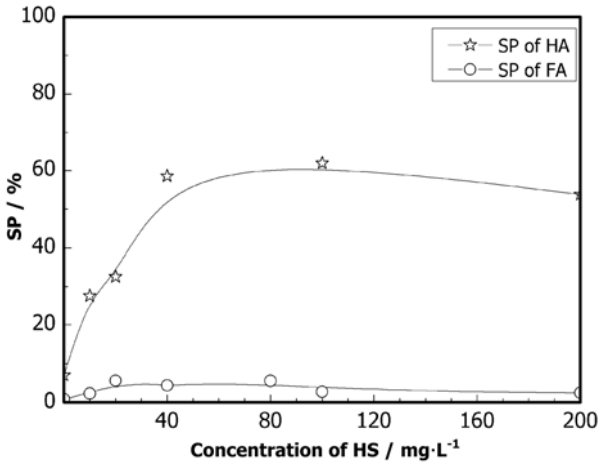

Fig.5 Effect of concentration of HS on uranyl humate precipitate. C_u=10 mg/L, I=0.1 mol/L, pH=6 and T=30℃.

3.2.5 Effect of ion strength

SO_4^{2-} was used to adjust ion strength in this experiment. Uranium precipitate was influenced evidently by ion strength from 0 mol/L to 0.5 mol/L

chinaXiv:202306.00189v1

ChinaXiv合作期刊

and kept 40%～50% of HA precipitate proportion. The data are plotted in Fig.6.

3.3 Migration of uranium in soil

Nuclide migration is one of the key issues for the radioactive contaminant in environment and affected by many factors. In this paper, the main aim is to study the influence of different humic substances on uranium migration in soil. Moreover, in order to understand the different effects between organic and inorganic colloid on migration, $Fe(OH)_3$ colloid solution and montmorillonite colloid were invesitigated. So FA, extracted HA (EHA), purchased HA (PHA), $Fe(OH)_3$ colloid solution, montmorillonite colloid mixed with original soil in proportion, as well as original soil were all tested at the same time.

Before uranyl ion was introduced into migration column, three columns were randomly chosen from the total samples for determination of water flow velocity with ^3H (Fig.7). The three outflow curves coincided exactly and the outflow velocity according to the curve could be calculated using a method developed by Liu, *et al*.[33] In this experiment condition, the average water flow velocity is calculated as 1.39 cm/L.

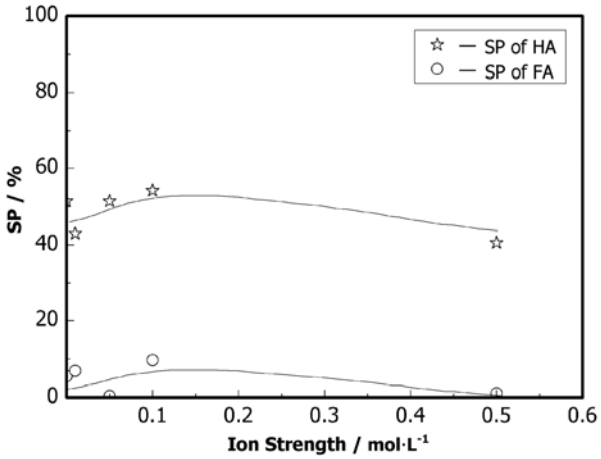

Fig.6 Effect of ion strength on precipitate of uranyl humate C_U=10 mg/L, C_{HA}=40 mg/L, C_{FA}=100 mg/L, pH=6, T=30℃.

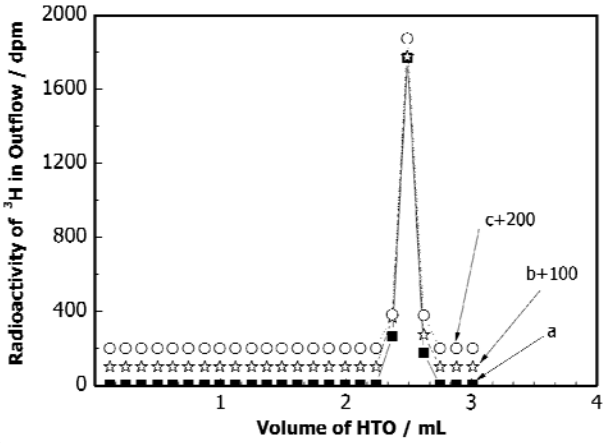

Fig.7 Outflow curve of HTO in migration columns. a, b and c were from random three migration columns, respectively. b and c have been shifted upward for clarity by plus counts indicated in the parentheses.

Table 3 Result of migration of U(VI) in different media

Characteristic of column	Volume of groundwater / mL	Retardation factors(Rd) / 10^4	Distribution ratios / 10^3 mL·g^{-1}	Migration distances / cm
Original soil	3527	1.0	2.2	2.0
Soil + additional FA	3720	0.9	2.1	2.2
Soil + additional EHA	3915	1.2	2.7	1.7
Soil + PHA	3845	1.3	3.1	1.5
Soil + $Fe(OH)_3$ colloid	3510	0.6	1.4	3.2
Soil + montmorillonite	3635	0.5	1.2	4.2

The curves about uranium migration in soil containing different colloids are depicted in Figs.8a-f. And the parameters of migration are reported in Table 3. According to the migration data, it could be noted that the additional FA could accelerate the velocity of uranyl ion migration, but EHA and PHA would retard the velocity when compared with uranyl in original soil under this experimental condition. However, despite the fact that FA not EHA or FHA can take uranyl far from the point of origin, the combination of HA and uranium was still limited and most of uranyl humate stayed at original point. The retardation by EHA or PHA could be either related to the adsorption described in our previous paper[32] or complexation of HA. The uranyl humate group formed in two reactions was too large to pass through the soil pore freely.

chinaXiv:202306.00189v1

ChinaXiv合作期刊

While $Fe(OH)_3$ and montmorillonite would rather accelerate the migration of uranyl because of its microne complex compound compared with the effect of inorganic colloid on the migration of uranyl in soil. In general, additional HA almost has no effect on the migration of U(VI) rather than inorganic colloid in soil.

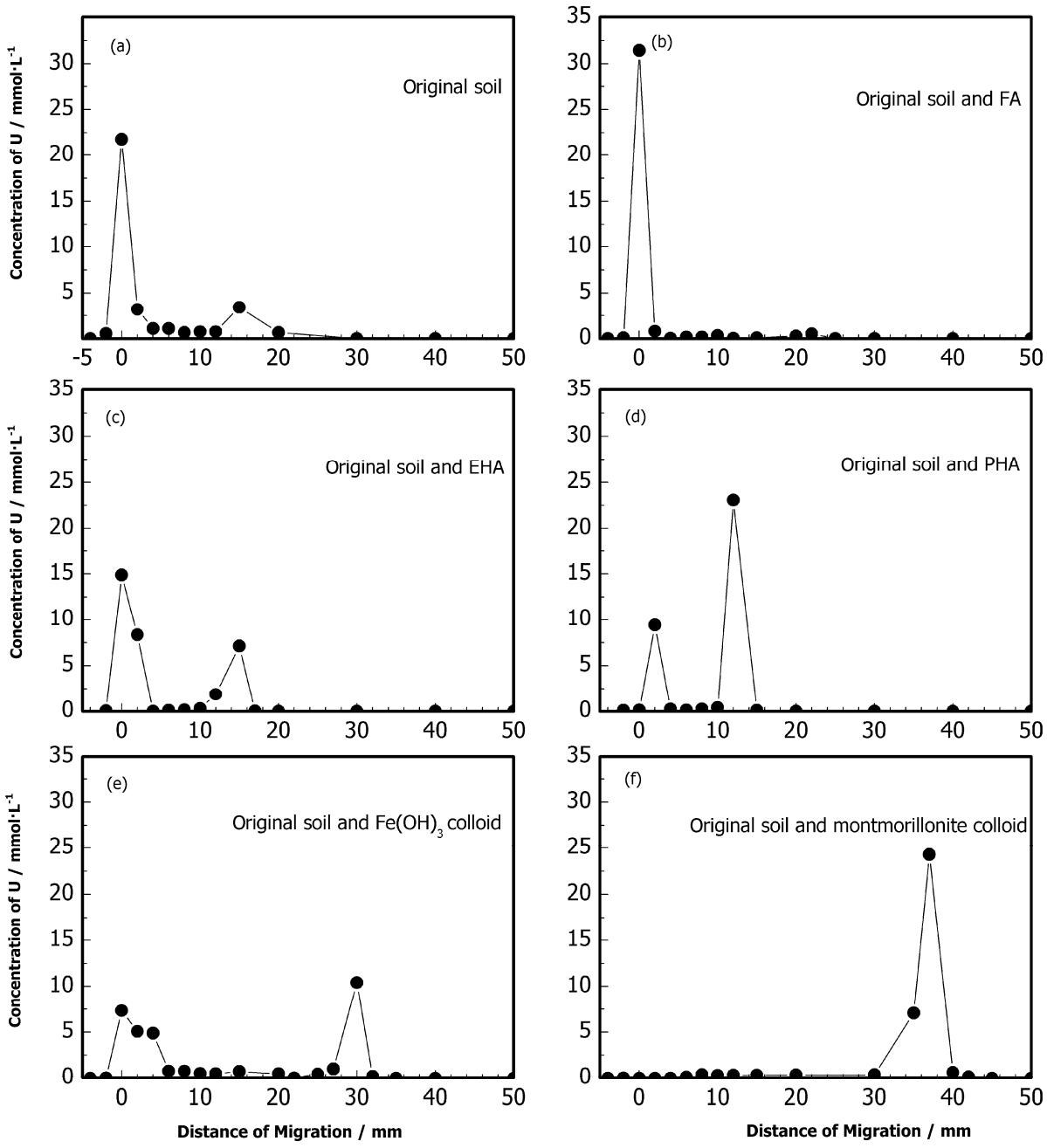

Fig. 8 Migration of U(VI) in different soil, (a) original soil, (b) original soil and FA, (c) original soil and EHA, (d) original soil and PHA, (e) original soil and $Fe(OH)_3$ colloid and (f) original soil and montmorillonite colloid.

4 Conclusion

Humic substance could complex with UO_2^{2+}, but the reaction was influenced by pH and temperature, and the complex compound was unstable. In different conditions, UO_2^{2+} could complex with HS in different ligand number as 1:1 or 1:2 or more.

We also found that, influenced by humic substance, UO_2^{2+} could be settled in different speciation in soil. But in the condition of 40 mg/L

chinaXiv:202306.00189v1

ChinaXiv合作期刊

uranium solution at pH 6 (I=0.1 mol/L, T=30℃), approximate 60% of precipitate was retarded by 0.45 μm micropore membrane, although it is much smaller than that of natural environment soil (The pore size of natural soil is around 1 μm). Moreover, the precipitation behavior was also influenced by pH, the concentration of uranium or HA as well as temperature in orderly.

In short, as the adsorption and complexation of uranium with HS, the behavior of UO_2^{2+} in soil containing natural organic material (NOM) was influence by additional HS. But this effect was limited and it could not change the migration state in generally. For the effect of other factors on uranium migration in soil will be further investigated in the future works.

Acknowledgements

The authors thank Dr. Jun Liao (Tissue Bioengineering Laboratory, Department of Agricultural and Biological Engineering, Mississippi State University, USA) for his help on the manuscript writing.

References

1 Andersson K, Evans S, Albinsson Y. Radiochim Acta, 1992, **58/59:** 321–327.
2 Borje T. Radiochim Acta, 1986, **39:** 105–112.
3 Pollner L, Zagyvai P. Waste Manage, 1999, **19**: 45–54.
4 Burnett W C, Elzerman A W. J Environ Radioactiv, 2001, **54:** 27–51.
5 Odintsov A A, Sazhenyuk A D, Satsyuk V A. Radiochemistry, 2004, **46:** 95–101.
6 Crancon P and van der Lee J. Radiochim Acta, 2003, **91:** 673-679.
7 Artinger R, Marquardt C M, Kim J I, *et al.* Radiochim Acta, 2000, **88**: 609–612.
8 Choppin G R. Radiochim Acta, 1988, **44/45:** 23–28.
9 Janos P. J Chromatogr A, 2003, **983:** 1–18.
10 Ran Y, Fu J, Rate A W, *et al.* Chem Geol, 2002, **185:** 33– 49.
11 Kim J I, Buckau G, Bryant E, *et al.* Radiochim Acta, 1989, **48:** 135–143.
12 Hummel W, Glaus M A, Van Loon L R. Radiochim Acta, 1999, **84:** 111–114.
13 Czerwinski K R, Buckau G, Scherbaum F, *et al.* Radiochim Acta, 1994, **65:** 111–119.
14 Higgo J J W, Kinniburgh D, Smith B, *et al.* Radiochim Acta, 1993, **61:** 91–103.
15 Caceci M S. Radiochim Acta, 1985, **39:** 51–56.
16 Kribek B, Podlaha J. Org Geochem, 1980, **2:** 93–97.
17 Rabung T, Geckeis H. Radiochim Acta, 2009, **97:** 265–271.
18 Struyk Z, Sposito G. Geoderma, 2001, **102:** 329–346.
19 Montavon G, Mansel A, Seibert A, *et al.* Radiochim Acta, 2000, **88:** 17.
20 Seibert A, Mansel A, Marquardt M C, *et al.* Radiochim Acta, 2001, **89:** 505.
21 Tao Z, Du J. Radiochim Acta, 1994, **64:** 225–228.
22 Jain A, Yadav K, Mohapatra M, *et al.* Spectrochim Acta A, 2009, **72:** 1122–1126.
23 Kim J I, Rhee D S, wimmer H, *et al.* Radiochim Acta, 1993, **62:** 35–43.
24 Tipping E. Radiochim Acta, 1993, **62:** 141–152.
25 Seijo M, Ulrich S, Filella M, *et al.* Environ Sci Technol, 2009, **43:** 7265–7269.
26 Kim J I. Radiochim Acta, 1991, **52/53:** 71–81.
27 Moulin V, Tits J, Ouzounian G. Radiochim Acta, 1992, **58/59:** 179–190.
28 Plavsic M, Kwokal Z, Strmecki S, *et al.* Fresen Environ Bull, 2009, **18:** 327–334.
29 Yang R J, van den Berg C M G. Environ Sci Technol, 2009, **43:** 7192–7197.
30 Andersen N P R, Mikkelsen L H, Keiding K. J Chin Inst Chem Eng, 2001, **32:** 525–528.
31 Sheng G P, Zhang M L, Yu H Q. J Colloid Interf Sci, 2009, **331:** 15–20.
32 Wei M, Liao J L, Liu N, *et al.* Nucl Sci Tech, 2007, **18:** 287–293.
33 Liu Q F, Liao J L, Liu N, *et al.* J Radioanal Nucl Ch, 2007, **274:** 593–601.

chinaXiv:202306.00189v1